cc: Chumbler/Seltzer
11/04/87

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October 9, 1987

Mr. J. W. Parks, Director
Enriching Operations Division
Department of Energy
Oak Ridge Operations
Post Office Box E
Oak Ridge, Tennessee 37831

Dear Mr. Parks:

Exposure Assessment - Paducah Feed Plant

The enclosed report provides the best estimate of radiation doses received by workers in the feed plant from transuranics (TRU) and fission products during the processing of uranium recycle materials. This information was requested as Item 6 of H. D. Fletcher's request of July 9, 1985. Available external exposure records and air monitoring data were used to arrive at these estimates. Scant internal dose data began to be generated by in vivo gamma spectrometry near the end of the recycle uranium processing.

Dose conversion factors derived by ICRP-30 Part 1 methods and provided by a DOE draft report were used to determine internal exposures. The transuranic nuclides ^{239}Pu and ^{237}Np provided about 12 percent of the 1.54 rem average annual combined total external dose and 50-year committed dose equivalent.

It is well known that the presence of ^{237}Np and ^{99}Tc in the recycle uranium processed at the Paducah Gaseous Diffusion Plant (PGDP) and in the UF_6 fed to the cascades has caused many radiation protection problems, especially during maintenance and modifications of process equipment related to the Cascade Improvement Program. Trapping technologies to remove these radionuclides were developed after the cascades were contaminated. Thus, future recycle feed streams can be cleaned up and cascade contamination can be avoided.

Also enclosed is a table listing the quantities and material form of uranium recycle material processed in the Feed Plant (per Item 1 of H. D. Fletcher's request). For Item 2, data are presented on the level of TRU and fission products in this recyle material in KY/L-1239, "Historical Impact of Reactor Tails in the Paducah Cascade," by R. F. Smith (3/19/84).

Sincerely,

ORIGINAL SIGNED BY
J. M. [illegible]

D. J. Bostock, Manager
Paducah Gaseous Diffusion Plant

DJB:MBrink

Enclosures

cc: K. W. Sommerfeld - ORGDP

cc/enc: D. L. Chumbler/S. F. Seltzer - HC
File (DJB) - -

RT, ERT, & SR-UO_3, U_3O_8, & CALCINED UO_3 FED IN C-420 & C-420

Year	Standard Tons U Fed	
1952	0.00	
1953	993.05	Start-up July 1953
1954	2,632.11	Includes 13.56 U_3O_8
1955	6,771.72	Includes 65.16 U_3O_8
1956	5,913.94	Includes 33.20 U_3O_8 & 1,825.53 Calcined UO_3
1957	6,564.75	Includes 39.97 U_3O_8 & 6,524.78 Calcined UO_3
1958	6,628.42	
1959	4,268.16	
1960	6,197.40	
1961	6,428.19	
1962	7,427.15	Includes 286.38 C-400 Calcined UO_3
1963	8,048.13	
1964	2,819.35	Shutdown June 1964
1965	0.00	
1966	0.00	
1967	0.00	
1968	3,496.03	Start-up July 1968
1969	8,032.25	
1970	2,130.89	
1971	5,457.73	
1972	5,359.46	
1973	4,028.14	
1974	0.00	
1975	0.00	
1976	0.00	
1977	364.77	Shutdown June 1977 Includes 45.71 Calcined UO_3
1978	0.00	
1979	0.00	
1980	0.00	
1981	0.00	
1982	0.00	
1983	0.00	
1984	0.00	
1985	0.00	

EXPOSURE ASSESSMENT - URANIUM RECYCLE MATERIALS IN THE PADUCAH FEED PLANT

R. C. Baker

One of the factors to be considered for recycling of irradiated uranium is the exposure of personnel during the conversion of uranium to uranium hexafluoride. The Paducah Feed Plant processed 100,000 tons of recycle uranium during the 1953-1973 period. The DOE-ORO requested the Paducah Gaseous Diffusion Plant and others to assess the exposure to transuranics and fission products (TRU and Fp's) for those employees processing this material. None of the organizations had readily accessible records or sufficient data to provide a timely assessment. This report provides an estimate of the exposure of employee groups with emphasis on the calculated incremental exposure due to the presence of the trace concentrations of TRU and Fp's in the returned

SUMMARY

From old records and interviews, estimates were made of 20-year average air concentrations and the particle size of the radioactive aerosols in the various operations areas and of the number of hours operators spent in each job. The fraction of the airborne radioactivity due to the various radionuclides was estimated for each area. Deposition factors were taken from Figure 5.1 of ICRP Part 1 to provide a correction for particle size. External radiation exposure was determined by film badge dosimetry. From this the average annual 50-year committed dose equivalent was calculated to be 1.54 [illegible] contributed 12 percent.

DESCRIPTION OF INPUT MATERIAL

The Paducah Feed Plant processed recycle uranium (RU) from July 1953 through June 1964 and from July 1968 through 1973. A small amount, 365 tons, was processed in early 1977. The feed plant has not been operated since June 1977.

The RU processed came from the plutonium production reactors at Hanford and Savannah River. It has been estimated that a total of 100,000 tons of uranium was processed and this contained 660 kg (11,000 curies) of ^{99}Tc, 18 kg (12 curies) of ^{237}Np and 330 grams (20 curies) of ^{239}Pu.

Fission product gamma activity was consistently well below 10 percent of that from aged natural uranium. However, ^{95}Zr-Nb and ^{106}Ru-Rh were detectable in the UO_3 feed. After concentration in dust following the fluorination process, ^{144}Ce was also detectable. The contribution of Fp gamma to the total radiation was usually small compared to that of uranium daughters. In 1957, it was noted that drums of fluorination system ash (unreacted uranium and nonvolatile fluorides) which were stored for over eight months (10 half-lives of ^{234}Th) had two to five times more gamma radiation than that expected from natural uranium. (The total gamma radiation level was much lower than that measured before storage.) Under this combination of concentration and aging, fission products can become the primary source of radiation. However, this situation was rare and the total dose from fission products was insignificant compared to that from other gamma emitters.

PROCESS DESCRIPTION

The simplified feed plant process used the following reactions:

UO_3 (yellow powder) + H_2 (gas)—> UO_2 (black powder) + H_2O (steam)

UO_2 + 4HF (gas) --> UF_4 (green powder) + $2H_2O$ (steam)

UF_4 + F_2 (gas) --> UF_6 gas

Impurities in the incoming UO_3 generally stayed with the solid process material until the final fluorination step. There were varying degrees of concentration of these impurities in the ash receivers, cooling lines, sintered metal filters and in the UF_6 product flow. When the conversion of UF_4 to UF_6 was exceptionally efficient, the residue was particularly concentrated. The radioactive decay products of ^{238}U when concentrated would emit gamma radiation measuring greater than 1000 mrem/h near the ash receivers (residue collectors). This material would also contain ^{239}Pu at 10 to 250 times the concentration in the feed material. (4 ppb in UO_3 X 250 = 1000 ppb or 1 ppm.)

When the efficiency of conversion was low and the residue (ash) was primarily unreacted UF_4, the radioactivity was low. Such ash was pulverized and screened in another facility and returned to the process at the fluorination system. Normal procedure for several years was to store the less pure ash for six months to a year to allow the more radioactive isotopes (such as ^{234}Th) to decay, dissolve the ash in nitric acid and chemically separate the uranium from the other isotopes. The uranium was then converted to UO_3 for refeeding [illegible] the residue from the [illegible] was discontinued in 1970.)

EMPLOYEE ASSIGNMENTS

Feed plant operators were assigned to day work or rotating shifts. In both cases they were rotated through several specific jobs as briefly described below. The potential for exposure was different for each job but, because of the job rotation, the operators had comparable exposure over an extended period.

1. UO_3 Powder Handling. This was primarily a day shift job involving two or three operators. It included unloading hoppers from railcars or trucks, weighing, valve installation, and transporting to the feed point. Exposure was about 220 hours per year to 80 uranium alpha dpm/m^3; particle Activity Median Aerodynamic Diameter (AMAD) was about 10 micrometers (um).

2. C-420 Green Salt Plant Operation. The normal personnel complement consisted of two day shift and three rotating shift operators. They manned the control room, checked screw reactors and valves, leak tested, unplugged feed screws and hoppers, cleaned-up spills, operated conveyors and baghouse dust collectors. Their typical annual exposure was 180 hours to 100 alpha dpm/m^3 and an additional 150 hours to 10 alpha dpm/m^3. The dust AMAD was approximately 10 um.

3. Fluorination Tower Operator. Normally, two operators on shift operated this area including checking the UF_4 hoppers and metering feed screws to the towers, F_2 preheater and control valve, ash receiver, off gas dust collectors, ventilation system, and transporting of filled ash receivers from the area. As much of the operation was done from the control room as was feasible. Unplugging off gas cooling lines, removing dust from piping and equipment, and clean up of spills were commonplace. There was exposure to

UO_2F_2 fume, gamma radiation from concentrated uranium daughters, and to dusts consisting of unreacted uranium containing enhanced concentrations of nonvolatile fluorides of metallic impurities including thorium, neptunium, plutonium and fission products. The UO_2F_2 fume had an AMAD of about 1 um; airborne dust from clean out operations had an AMAD of about 3.5 to 7 um. An AMAD of 4 um is assumed for exposure calculations. The annual exposure time was 100 hours to UO_2F_2 concentrations of 150 alpha dpm/m^3 and 120 hours to other dust at 80 alpha dpm/m^3.

4. Cold Trap and Refrigeration Systems. In addition to operating these systems, the operator would connect and disconnect UF_6 cylinders and move and weigh cylinders. This work rarely generated air contamination, but there were a few instances of acute exposure to UO_2F_2 from releases of UF_6. There was also chronic exposure to dust generated in other areas. The estimated annual dust exposure was 240 hours to 20 alpha dpm/m^3 (AMAD estimated at 4 um). Some operators were exposed to one or more releases in which 1 to 2 mg of soluble uranium in UO_2F_2 was inhaled.

5. C-410 Control Room Operation. Much of the process was instrumented and controlled remotely with the operating conditions displayed on graphic panel boards. The control room operator monitored the displays, communicated with the local operator, and occasionally went to his assistance. The control room, an area of no significant exposure, was also used as the assigned work area for personnel who were placed on work restriction when their uranium excretion rates exceeded a plant action guide. The estimated annual exposure was 320 hours at an average airborne alpha emitter concentration of 10 cpm/m^3.

6. Fluorine Plant Operation. There was essentially no radiation exposure to the personnel operating or maintaining the fluorine plant or the HF tank farm. In their normal rotation, the operators spent about 300 hours per year on this job. Operators whose urinalyses indicated excretion rates above the plant action guides may be assigned to this job until the rates were within the guides.

During the period of 1953-1960, feed plant personnel operated a facility to vaporize UF_6 to the cascade. There was no exposure to TRU or Fp's in this operation.

NUCLIDES IN FEED PLANT AEROSOLS

Data from a few samples of dust in process systems and the many analyses of feed materials were used with material balance data to estimate the TRU and ^{230}Th content of dust in the various work areas of the feed plant. These concentrations are used to calculate the annual intake of each isotope.

1. UO_3 Powder Area.

Isotope	Conc.	dpm/gU	Fraction of Total Dust Alpha Count
^{239}Pu	3.3 ppb	500	3.3×10^{-4}
^{237}Np	0.18 ppm	270	1.8×10^{-4}
^{238}U	0.993 g/g	750,000	0.5
^{234}U	56 ppm	750,000	0.5
^{230}Th	1 ppb	42,000	2.8×10^{-5}
		1,500,818	

2. C-420 Green Salt Plant

Same as for UO_3 Powder Area

3. Fluorination Tower Area. TRU concentrations varied greatly with location and operating conditions. The TRU concentrations were very low when malfunctions or poor conversion efficiency resulted in excessive dust in the off gas system. Good operation resulted in higher concentrations but less need to open the system. Systems with long trouble-free operation had high external radiation, high concentration of thorium isotopes and enhanced TRU. The values given below are used to represent the wide range of nuclide concentration in the airborne dust.

Isotope	Conc. (U basis)	dpm/g	Fraction of Total Dust Alpha Count
^{239}Pu	1 ppm	150,000	0.09
^{237}Np	8 ppm	12,000	.007
^{238}U	0.99 g/g	750,000	0.44
^{234}U	56 ppm	750,000	0.44
^{230}Th	1 ppm	42,000	0.025
		1,704,000	

UO_2F_2 fume in tower area had essentially no TRU or ^{230}Th content.

The above distribution of radionuclides is considered typical. Dust from components rarely opened may contain greatly different distributions. For example, dust was sampled from scintered metal filters through which the UF_6 gas and excess fluorine had passed during 18 months of use on some RU but primarily natural uranium. Of the total alpha activity of the dust, ^{230}Th alphas constituted 80 percent; ^{239}Pu was less than 4 percent and ^{237}Np was less than 1 percent of the total activity.

4. Cold Trap and Refrigeration Operation.

Exposure was to UO_2F_2, dust drifting from the tower area and dust from UO_3 area. It is estimated that 80 percent of the dust came from the tower area and 20 percent from UO_3. Thus, the isotopic distribution would be as follows:

Isotope	Conc. (U basis)	dpm/g	Dust Count
^{239}Pu	0.8 ppm	120,000	0.07
^{237}Np	0.6 ppm	9,000	0.005
^{238}U	0.99 g/g	750,000	0.45
^{234}U	55 ppm	750,000	0.45
^{230}Th	0.8 ppm	34,000	0.02
		1,663,000	

5. C-410 Control Room Operations.

The dust to which control room operators were exposed is assumed to have the same nuclide distribution as that for tower operators.

RADIONUCLIDE INTAKE ESTIMATES BASED ON ANNUALIZED AIR CONCENTRATIONS

An inhalation rate of 1.2 cubic meter per hour is assumed for the moderate work rate. The total activity inhaled and deposited in the pulmonary region of employee lungs is calculated below:

PLUTONIUM AND NEPTUNIUM AND URANIUM DEPOSITION

Job	Time Hours	Alpha dpm/m^3	Resp. Rate M^3/Hr	Nuclide Fraction	Deposition* Factor	Total Annual Deposits (dpm)
(1)	220	80	1.2	Pu 3.3×10^{-4}	0.05	0.36
				Np 1.8×10^{-4}	0.05	0.19
				U 1.0	0.05	1×10^3
(2)	180	100	1.2	Pu 3.3×10^{-4}	0.05	0.38
	150	10		Np 1.8×10^{-4}	0.05	0.22
				U 1.0	0.05	1.1×10^3
(3)	100	150	1.2	U 1.0	0.25	4.5×10^3
	120	80	1.2	Pu 0.09	0.1	104
				Np 0.007	0.1	8.1
				U 0.9	0.1	1.0×10^3
(4)	240	20	1.2	Pu 0.07	0.1	40
				Np 0.005	0.1	2.8
				U 0.94	0.1	0.5×10^3
(5)	320	10	1.2	Pu 0.09	0.1	35
				Np 0.007	0.1	2.6
				U 0.9	0.1	0.3×10^3
(6)	300	0	1.2	---	—	—

*Deposition factor is the fraction of inhaled particulate mass which reaches and is deposited in the lungs.

Total 1,630 hours — Total Annual Deposit (dpm)$_3$ — Pu 180 dpm, Np 13.9 dpm
Nonexposure hours 370 — Uranium Class W 3.9×10^3
Uranium Class D 4.5×20^3

These total annual deposits may be compared to Annual Limits of Intake (ALI) by dividing by 0.25, the assigned deposition factor for dust with an AMAD of 1 micrometer.

^{237}Pu	720
^{237}Np	56
U (D)	[illegible]
U (W)	[illegible]

FISSION PRODUCT EXPOSURE

Recycle uranium received at the Paducah feed plant typically contained 7 ppm ^{99}Tc on a uranium basis. It is estimated that 660 Kg ^{99}Tc was received in 100,000 tons of uranium. Technetium did not concentrate in any particular part of the feed plant; the input concentration was typical of that in airborne dust. Technetium accumulated in the cascade and in cylinder heels. A concentration of 7 ppm results in 2.6×10^5 technetium beta dpm/gram uranium. The airborne dust had ^{99}Tc beta activity of about 1/6 of the ^{234}Th-Pa beta activity, except where the ^{234}Th, concentrated by the process, was much greater. Annual deposition is estimated to be 1,000 dpm (16 Bq) or less than 0.05 mrem/year to lungs or other tissue.

As received the recycle uranium had fission product gamma activity of less than one tenth the gamma activity of aged natural uranium. Normally specific nuclides were not identifiable. However, concentration of fission products in ash made ^{106}Ru-Rh, and ^{95}Zr-Nb detectable. Gamma dose rates in the tower area, around ash receivers and other tower area equipment whether installed, stored, or being cleaned was significant. Dose rates near or in contact with some equipment reached 1 rem/hr. Almost all the gamma radiation was from the uranium decay chain. The dose rate from equipment removed from the process diminished at the decay rate of ^{234}Th during the first months of storage. The gamma radiation due to ^{234}Th-Pa was probably about 20 or more times that due to fission products.

EXTERNAL EXPOSURE

Dosimetry records were reviewed for 27 operators who in over a period of 7 to 22 years of feed plant operation received most of their exposure in the feed plant. The average annual dose was 0.66 rem whole body and 2.7 rem skin dose. Of this total the TRU and fission produce contribution is estimated to be less than 20 mrem/year to the whole body. This is based on analyses which generally expressed the gamma from Fp's to be either "less than 2 percent of aged normal U" or "significantly less than 10 percent of aged normal U." Fission product beta activity, other than from ^{99}Tc, was reported as "less than 1 percent of aged normal U."

Dosimetry records of maintenance mechanics assigned to the feed plant were also reviewed. Their exposure was slightly higher than that for the operators. The whole body dose averaged 0.74 rem/year and the skin dose averaged 2.9 rem/year. Again, TRU and Fp's added very little, perhaps 20 mrem/year to the total.

IN VIVO MONITORING DATA

The early evaluation of the few analyses for TRU and Fp's in the feed plant confirmed the prediction that exposure to those radionuclides would be minor compared to exposure to uranium. However, the project to recover and purify neptunium and the discovery of significant deposition of ^{237}Np on cascade equipment made monitoring for Np necessary. While monitoring personnel performing maintenance on cascade equipment became the highest priority, there was some monitoring of feed plant personnel by in vivo gamma spectrometry (body counting) before the feed plant closed in 1977.

The earliest use of a "Body Counter" to check for internal deposition of radionuclides in Paducah Plant employees was in 1960 and primarily was because of concerns for exposure to ^{237}Np. At the request of PGDP, Y-12 calibrated their in vivo gamma spectrometer for ^{237}Np. It was found that good sensitivity was obtainable; less the 2 percent of the 17 nCi Maximum Permissible Lung Burden (MPLB) could be detected.

In 1960, four chemists who worked on the development of a neptunium recovery and purification system and were potentially exposed to ^{237}Np and ^{239}Pu were checked in the calibrated Y-12 in vivo spectrometer. No ^{237}Np deposits were found.

In 1962, 15 employees considered to be among those with the highest potential exposure to neptunium and uranium in each of three plant groups (^{237}Np recovery, converter disassembly, feed plant operations) were sent to Y-12 to be counted. None of the employees had detectable ^{237}Np.

In 1968, the mobile In Vivo Radiation Monitoring Laboratory (IVRML) began regular visits to PGDP. However, routine calibration and recording of readings for lung deposits of ^{237}Np were not begun until 1974, after almost all RU had been processed in C-410. Of 66 determinations for feed plant operators, the maximum indicated deposit was 0.5 nCi. This is considered to be 3 percent of the MPLB. The mean determination was not greater than that of unexposed controls.

EXPOSURE OF OTHER PLANT GROUPS TO TRU AND FISSION PRODUCTS

The importance of exposure to TRU alpha emitters was recognized during the cascade improvement program of the 1959-1962 era. All analyses, surface

contamination control, respiratory protection, and a urinalysis program were all used to assess and control exposure. While the air analyses predicted annual exposures to average near the radioactivity concentration guides, the IVRML lung counts showed low deposition (generally less than 2 percent of the MPLB). A summation of 25 years of monitoring indicates the following:

1. Plutonium and neptunium concentrate in UF_6 cylinder heels. It is not accessible until the cylinder is washed for inspection and pressure testing. The potential for personnel exposure is low during the wet chemical processing of cylinder wash solutions. Processes which would generate aerosols are avoided.

2. Neptunium in UF_6 feed to the cascade will contaminate the process equipment and its control must be considered during cascade maintenance or improvement. The level of neptunium contamination can be reduced to insignificance by the use of cobaltous fluoride traps in the feed line.

3. Deposition of ^{237}Np in the lung from exposure to dust from cascade equipment disassembly and modification was lower than predicted by air sampling because of the large size of the dispersed particulates.

4. The "body count" as done by the IVRML was the most direct and most reliable exposure monitoring method for ^{237}Np exposure. Since ^{237}Np was present whenever ^{239}Pu was found, a negative determination for ^{237}Np by in vivo chest counting was also an indication that there was no significant plutonium deposition. Except for feed plant

dusts where the TRU activity was low compared to uranium, the ^{237}Np activity in materials accessible to employees was normally 2 to 20 times that of ^{239}Pu.

DOSE SUMMARY

There are various systems to convert deposits or intakes of radionuclides to dose units. Conversion factors for this summary were taken from a draft table "50 Yr. Committed Dose Equivalent Factors - Rem/uCi Intake" issued by DOE in 1985 for population dose calculations. This was used in lieu of the supplement to ICRP-30 Part 1, which does not have values for ^{237}Np.

DOSE SUMMARY - FEED PLANT OPERATORS

Nuclide	Solubility Class	Intake uCi/Yr.	Conv. Factor REM/uCi	50 Year Committed Dose Equivalent (REM)
$^{234}U+^{238}U$	D	0.082	2.6	0.21
$^{234}U+^{238}U$	W	0.073	6.7	0.49
^{237}Np		2.5×10^{-5}	490	.012
^{239}Pu	W	3.2×10^{-4}	510	.17
Total Internal 50 Year Committed Dose Equivalent				0.88
External Dose (Film Badge gamma)				0.66
Total Dose (Annual Average)				1.54 REM/YR.

CONCLUSIONS

The radiation dose received by personnel during the conversion of 100,000 tons of recycled uranium averaged 1.5 rem per year including the committed dose equivalent from internally deposited radioisotopes. Most of the dose was due to the radioactivity of naturally occurring isotopes of uranium, about 12 percent was due to transuranics and fission products. The neptunium and technetium that passed through the feed plant and was fed to the gaseous diffusion plants added significantly to the radiation protection problems of the Cascade Improvement Programs. Most of this contamination was removed during these programs. The technology for removing plutonium, neptunium and technetium from UF_6 with chemical traps has been demonstrated and if used can effectively prevent recontamination of the cascades.